

Confinement and non-universality of anomalous heat transport and superdiffusion of energy in low-dimensional systems

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We provide molecular dynamics simulation of heat transport and thermal energy diffusion in one-dimensional molecular chains with different interparticle pair potentials at zero and non-zero temperature. We model the thermal conductivity (TC) and energy diffusion in the coupled rotator chain and in the Lennard-Jones chain either without or with the confining parabolic interatomic potential. The considered chains without the confining potential have normal TC and energy diffusion, while the corresponding chains with the confining potential are characterized by anomalous (diverging with the system length) TC and superdiffusion of energy. We confirm in such a way that, surprisingly, the confinement makes both heat transport and energy diffusion anomalous in low-dimensional phononic systems. We show that the chain, which has a finite TC, is also characterized by the normal energy diffusion in the thermalized chain (at non-zero temperature), while the superdiffusion of thermal energy occurs in the thermalized chains with only anomalous TC. We present the arguments, supported by our simulations, that the scaling relation between the exponents in time dependence of the mean square displacement of thermal energy distribution and in length dependence of the anomalous TC is not universal and can be different, depending on the main mechanism of energy transport: by weakly-scattered waves or by noninteracting colliding particles performing Lévy flights.

Phonon thermal conductivity in low-dimensional systems and at nanoscale presents a challenge both from theoretical and experimental points of view [1, 2]. One of important problems in this field is the origin of anomalous, diverging with the system size, thermal conductivity (TC) of one-dimensional (1D) lattice models [3–10] and quasi-1D nanostructures and polymers [11–13]. The length-dependent TC was experimentally observed in carbon nanotubes [14] and in suspended single-layer graphene [15]. The general belief is that TC is anomalous in 1D momentum-conserving systems [16]. Nevertheless, a chain of coupled classical rotators presents an example of translationally-invariant (isolated) and correspondingly momentum-conserving 1D periodic system with finite TC [17, 18]. In a recent paper [19], normal heat transport was reported for 1D momentum-conserving systems with the Lennard-Jones, Morse, and Coulomb potential. It was shown in [19] that the convergence of TC is provided by phonon scattering on the locally strongly stretched loose interatomic bonds at low temperature and by the many-particle scattering at high temperature.

While the research on TC in low-dimensional systems is very intensive, the peculiarities of energy diffusion have been much less studied. Usually the nonthermalized chain has been considered with only small central portion being thermalized, and thermal energy propagation in the chain was studied afterwards. Such modeling can lead to anomalous (super-) diffusion of the thermal energy, and sometimes it is concluded that such 1D system possesses anomalous thermal conductivity [20]. For example, superdiffusion of energy was obtained in Ref. [20] in the chain with the Lennard-Jones (LJ) interparticle potential, in which the normal heat transport has been revealed previously [19, 21]. This suggests the incorrectness

of the simulations of energy diffusion because the latter was simulated in Ref. [20] in the chain at zero background temperature. Any anharmonic chain at low temperature behaves as an almost harmonic chain, in which the heat transport is carried out by ballistic phonons and TC diverges with the chain length. Therefore it is not surprising that in the nonthermalized chain (at zero background temperature) the energy propagates ballistically. But this does not imply that the superdiffusion of energy will also hold in the thermalized chain (at non-zero background temperature).

On the other hand, the chains with a (parabolic or quartic) confining pair potential, which does not allow for bond dissociation, are expected to possess anomalous thermal conductivity, diverging with the chain length [19]. The celebrated Fermi-Pasta-Ulam potential belongs to such type of the pair potentials. In this paper, we model both the TC and diffusion of energy in anharmonic chains at zero and non-zero temperature. We show that the chain, which has a finite TC, is also characterized by the normal diffusion of energy in the thermalized chain, while the anomalous superdiffusion of thermal energy occurs in the thermalized chains with only anomalous (diverging with the system length) TC. To show this, we model TC and energy diffusion in the coupled rotator chain (a chain with periodic interatomic potential) and in LJ chain either without or with the parabolic confining pair potential. Such parabolic confining pair potential is introduced, for instance, on top of the shallow LJ potential to hold together the constituent cellular particles within a cell [22]. The considered chains without the confining potential are characterized by the normal diffusion of energy in the thermalized chain, while the corresponding chains with the confining pair potential are characterized by the superdiffusion of energy. We confirm in such a way that, surprisingly, the confinement makes both heat transport and energy diffusion anomalous in low-dimensional systems. We present the arguments, supported by our simulations, that the scaling relation between the exponents in time dependence of the mean square displacement

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of thermal energy distribution and in length dependence of the TC, which is discussed in connection with anomalous heat transport and superdiffusion of energy in low-dimensional systems, see, e.g., Refs. [23–27], is not universal and can be different, depending on the main mechanism of energy transport – either by weakly-scattered waves or by noninteracting colliding particles performing Lévy flights.

We consider the periodic molecular chain consisting of $N = L/a$ unit cells, where L is the chain length, a is a lattice period. In a dimensionless form, the Hamiltonian of the chain can be written as

$$H = \sum_{n=0}^N \frac{1}{2} \dot{x}_n^2 + \sum_{n=0}^{N-1} V(x_{n+1} - x_n), \quad (1)$$

where x_n is the displacement on the n th particle from its equilibrium position at na , $V(r)$ is a dimensionless pair interaction potential between nearest neighbors normalized by the conditions $V(0) = 0$ and $V'(0) = 0$.

To simulate the heat transfer in the chain, we use the stochastic Langevin thermostat. We consider a finite chain of $N_+ + N + N_-$ unit cells, and take the chain with fixed ends. We put the N_- left boundary particles in the Langevin thermostat with temperature T_- , and N_+ particles in the right-hand edge Langevin thermostat with a temperature of T_+ . The corresponding system of equations of motion of the chain is:

$$\begin{aligned} \ddot{x}_n &= -\partial H / \partial x_n - \gamma \dot{x}_n + \xi_n^+, \quad n \leq N_+, \\ \ddot{x}_n &= -\partial H / \partial x_n, \quad n = N_+ + 1, \dots, N_+ + N, \\ \ddot{x}_n &= -\partial H / \partial x_n - \gamma \dot{x}_n + \xi_n^-, \quad n > N_+ + N, \end{aligned} \quad (2)$$

where γ is relaxation coefficient of the particle velocity, ξ_n^\pm are random forces which simulate the interaction with the white-noise thermostat normalized by the conditions

$$\langle \xi_n^\pm(t) \rangle = 0, \quad \langle \xi_n^\pm(t_1) \xi_k^\pm(t_2) \rangle = 2\gamma T_\pm \delta_{nk} \delta(t_2 - t_1).$$

We integrate numerically the Langevin equations of motion (2) by employing the Verlet velocity method with the step $\Delta t = 0.02$. After some integration time t_0 (this value depends on the chain length between the thermostats), we observe the formation of a temperature gradient and constat heat energy flux in the central part of the chain. After the stationary heat flux is established, we can find the temperature distribution by using the relations for $T_n = \langle \dot{x}_n^2 \rangle_t$ and stationary heat flow along the chain $J_n = -\langle \dot{x}_n V'(x_n - x_{n-1}) \rangle_t$. The following values were used in the numerical simulation: $T_\pm = (1 \pm 0.1)T$, $\gamma = 0.1$, $N_\pm = 40$, $N = 20, 40, 80, \dots, 20480$. The detailed description of the Langevin equations and justification of the method are presented in [19].

In the steady-state regime, the heat flux through each cell in the central part of the chain should be the same, i.e., $J_n \equiv J$, $N_- + 1 \leq n \leq N_- + N + 1$. Almost linear gradient of temperature distribution is established in the central part of the chain, so we can define TC as

$$\kappa(N) = J(N-1)/(T_{N_++1} - T_{N_++N}). \quad (3)$$

In our modeling of the TC, we use the following pair interaction potentials:

$$V(r) = 1 - \cos(r), \quad a = 2\pi, \quad (4)$$

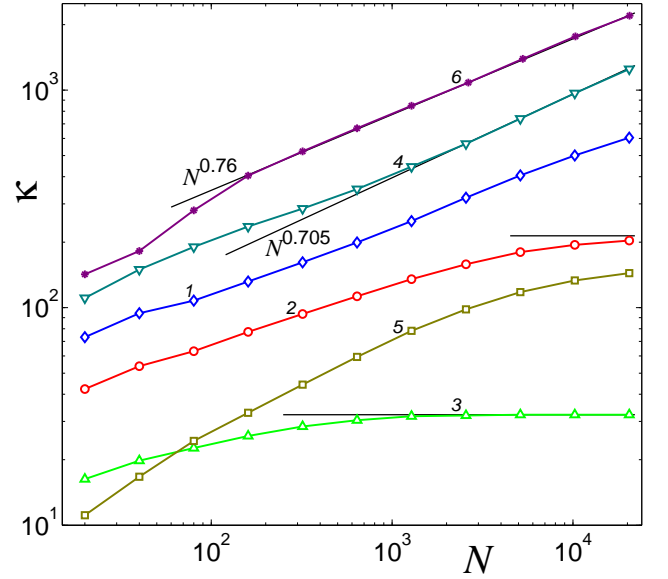


Figure 1: Thermal conductivity κ versus dimensionless chain length $N = L/a$ for the chain with the periodic potential (4) at normalized temperature $T = 0.15, 0.2, 0.3$ (curves 1, 2, 3), for the chain with the combined potential (5) at $T = 0.3$ (curve 4), for the chain at $T = 0.002$ with the Lennard-Jones potential (6) (curve 5, which shows for the convenience 0.15κ) or with the combined potential (7) (curve 6).

– the periodic potential,

$$V(r) = 1 - \cos(r) + 0.25r^2, \quad a = 2\pi, \quad (5)$$

– the sum of the periodic and parabolic confining potentials,

$$V(r) = 4\epsilon[(\sigma/(1+r))^6 - 1/2]^2, \quad a = 1, \sigma = 2^{-1/6}, \epsilon = 1/72, \quad (6)$$

– the LJ potential,

$$V(r) = 4\epsilon[(\sigma/(1+r))^6 - 1/2]^2 + 0.25r^2, \quad (7)$$

– the sum of the LJ and parabolic confining potentials. Note that one has $V''(0) = 1$ for potentials (4) and (6) and therefore speed of sound (small-amplitude phonons) in the chain is $v_s = a$, while for potentials (5) and (7) one has $V''(0) = 1.5$ and therefore $v_s = a\sqrt{1.5} = 1.2247a$.

Dependence of TC on dimensionless chain length $\kappa(N)$ is presented in Fig. 1. As one can see from this figure, $\kappa(N)$ in the chain with the periodic potential (4) depends on temperature: the convergence of $\kappa(N)$ for $N \rightarrow \infty$ is slower for lower temperature. Thermal conductivity saturates for $N = 2560$ at $T = 0.3$, for $N = 20480$ at $T = 0.2$, and there is no saturation of the $\kappa(N)$ at the maximal used $N = 20480$ at $T = 0.1$ (and one needs $N \sim 10^7$ to reach the saturation at such temperature). This feature is related with the increase of phonon mean free path with the decrease of temperature because the saturation of $\kappa(N)$ occurs only when the chain length $L = Na$ reaches or exceeds phonon mean free path.

The chain with the combined interatomic potentials (5) and (7) is characterized by anomalous heat transport. Here TC

monotonously diverges with N as $\kappa(N) \sim N^\alpha$. For the potential (5) at $T = 0.3$, the exponent $\alpha = 0.705$, for the potential (7) at $T = 0.002$, the exponent $\alpha = 0.76$. TC of the chain with the LJ potential (6) saturates for $N \sim 10^4$ at $T = 0.002$, as one can see in Fig. 1.

Now we analyze how the $\kappa(N)$ dependence is related with thermal energy propagation at finite temperature ($T_0 > 0$). To this end, we consider the chain with N atoms with the fixed ends. We thermalize the chain with the use of the Langevin heat bath such that the first N_l atoms have the temperature T_l , while the rest of the atoms have the temperature $T_0 < T_l$. We consider the Langevin equations of motion with random forces:

$$\ddot{x}_n = -\partial H / \partial x_n - \gamma \dot{x}_n + \xi_n, \quad 0 < n < N, \quad (8)$$

when $x_0 \equiv 0$, $x_N \equiv 0$, and the random forces ξ_n are δ -correlated as

$$\langle \xi_n(t_1) \xi_k(t_2) \rangle = 2\gamma T_n \delta_{nk} \delta(t_2 - t_1),$$

with $T_n = T_l$ for $n = 1, \dots, N_l$, and $T_n = T_0$ for $n = N_l + 1, \dots, N - 1$. Now we take the (classical) zero-energy initial condition $\{x_n(0) = 0, \dot{x}_n(0) = 0\}_{n=1}^{N-1}$ and integrate the Langevin equations (8) from $t = 0$ to $t_1 = 20/\gamma = 200$. In result we get a random realization of the initial state of the thermalized chain $\{x_n(t_1), \dot{x}_n(t_1)\}_{n=1}^{N-1}$ with $T = T_l$ in its left end and $t = T_0 < T_l$ in the rest of the chain.

Then we model the propagation of the thermal energy in the chain, which starts from its left end with higher temperature. To perform this, we integrate equations of motion of the atoms without their interaction with the thermostat,

$$\ddot{x}_n = -\partial H / \partial x_n, \quad 0 < n < N, \quad (9)$$

when $x_0 \equiv 0$, $x_N \equiv 0$, with the initial condition which were obtained after the integration of the Langevin equations of motion (8). In order to analyze the propagation of the thermal energy during the long time in the chain with $N = 20000$ atoms, we take the number of the end atoms $N_l = 40 \ll N$. We trace the time dependence of the distribution along the chain of the temperature $T_n(t) = \langle \dot{x}_n^2(t) \rangle$ and of the energy $E_n(t) = \langle \dot{x}_n^2(t)/2 + V(x_n(t) - x_{n-1}(t)) \rangle$, where the averaging is taken over the independent realizations of the initial thermalized state of the chain. Time evolution of the temperature distribution in the chain with the periodic potential is shown in Fig. 2. It is worth noting that for the analysis of thermal energy distribution in the thermalized chain, with $T_0 > 0$, one needs to perform the averaging over a large number ($\sim 10^5$) of independent realizations of the initial thermalized state [while for the zero-energy initial state, with $T_0 = 0$, the averaging can be performed over significantly lower number ($\sim 10^3$) of independent realizations of the initial state].

We consider first the chain with the periodic interatomic potential (4). The left end of the chain is thermalized at $T_l = 1$, and the rest of the chain – at T_0 , when $0 \leq T_0 < T_l$. As one can see in Fig. 2, the thermal energy distribution substantially depends on the chain temperature T_0 . For $T_0 = 0$, the thermal energy propagates ballistically, with the sound speed, along

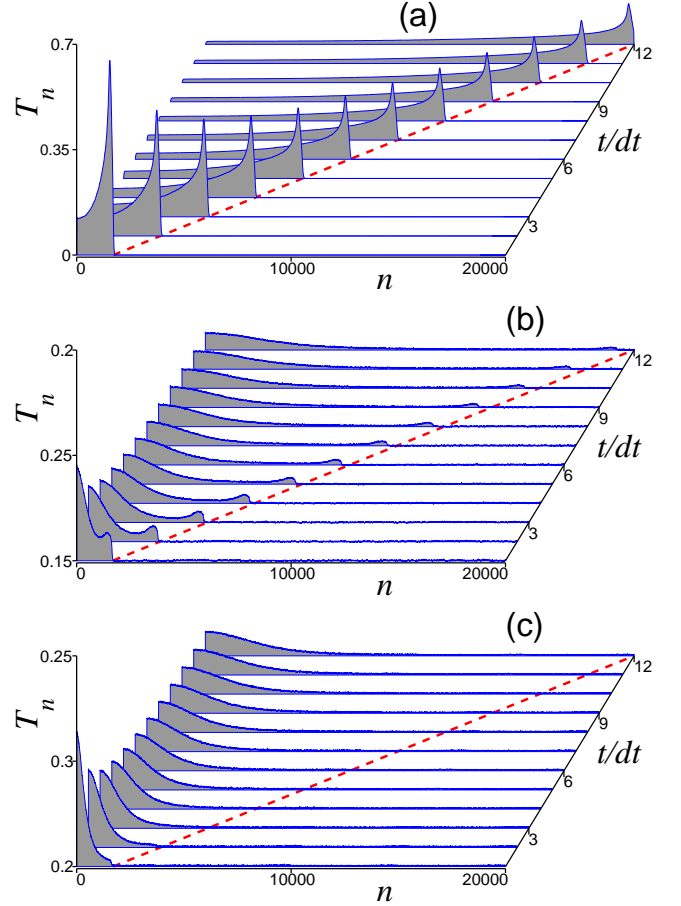


Figure 2: Propagation of temperature distribution T_n in a chain with the periodic potential (4) for $T_l = 1$ and $T_0 = 0$ (a), $T_0 = 0.15$ (b), $T_0 = 0.2$ (c). Temperature distributions are shown for characteristic delay time $dt = 1662.5$, dashed lines show sound cone $n = v_s t / a$.

the chain. While for $T_0 = 0.15$, only relatively small part of thermal energy propagates ballistically, which then dissipates in the chain. The main part of thermal energy is concentrated at the left, high-temperature, end of the chain and spreads slowly towards the right end. For the higher temperature of the chain, $T_0 = 0.2$, there is no ballistic energy propagation along the thermalized chain: the energy distribution spreads diffusively from the very beginning.

The unilateral spreading can be described by the following mean square displacement of thermal energy distribution (MSDTED) of the excess energy, initially placed in the site $n = 1$ of the chain with $u_0 \equiv 0$, $u_N \equiv 0$:

$$\langle \Delta x^2 \rangle(t) = \sum_{n=1}^{N-1} (n-1)^2 e_n(t). \quad (10)$$

Here $e_n(t) = (E_n(t) - E_0)/E$ is a discrete distribution of normalized excess energy in the chain, $E = \sum_{n=1}^{N-1} (E_n - E_0)$ is a constant total excess energy, $E_0 = T_0$ is the average particle energy in the thermalized chain at temperature T_0 , see

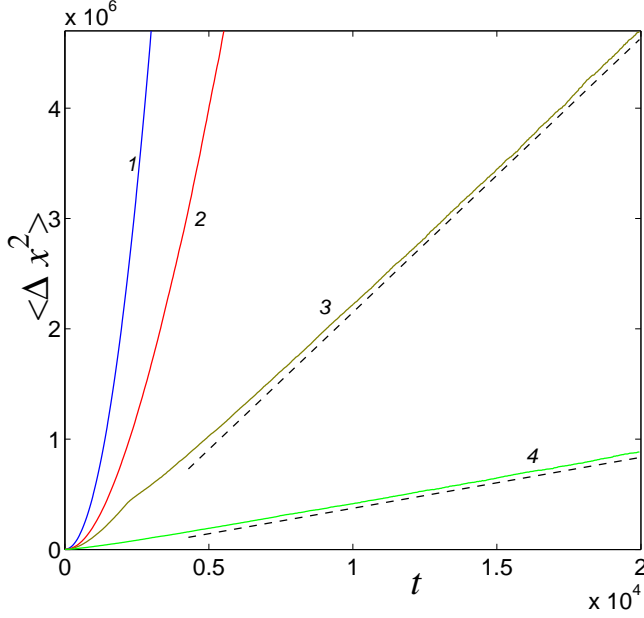


Figure 3: Time dependence of MSDTED, Eq. (10), in the chain with the periodic potential (4) for the chain temperature $T_0 = 0, 0.15, 0.2$, and 0.3 (curves 1, 2, 3, and 4). Dotted lines show the linear dependencies.

also Refs. [27, 28]. Mean square displacement can also be introduced for the discrete temperature distribution along the chain, when $T_n(t) = \langle \dot{x}_n^2(t) \rangle$, and both mean square displacements, of thermal energy and temperature distributions, have the same time dependence.

Time dependence of MSDTED in the chain with the periodic potential (4) is shown in Fig. 3. As one can see in this figure, at zero and low chain temperature, $T_0 = 0$ and $T_0 = 0.15$, MSDTED grows quadratically with time, $\langle \Delta x^2 \rangle \propto t^2$, which corresponds to the ballistic energy propagation. At higher temperature $T_0 = 0.2$, during the short delay time $t < 10^3$ MSDTED also grows quadratically with time, but for longer delay $t > 10^4$ MSDTED changes to become linear in time, which corresponds to the normal energy diffusion. As one can also see in Fig. 3, the normal energy diffusion starts earlier for the higher temperature of the chain which is a consequence of the enhanced phonon scattering by thermally-activated anharmonicity of the chain. From Fig. 3 follows that the used chain length N is not enough long to model the transition from the ballistic to normal diffusion regime of energy propagation at temperature $T_0 = 0.15$. We also note that this length of the chain is not enough to model the normal TC in it at this temperature as well. We can conclude from the comparison of Figs. 1 and 3 that the minimal chain length N_{min} , at which the normal TC is established, is directly related with the minimal delay time t_{min} , which is needed for the normal energy diffusion to be established in the same chain: $N_{min} \approx v_s t_{min} / a$.

Similar time dependence of MSDTED is revealed in the chain with the LJ interatomic potential, see Figs. 4 and 5. We

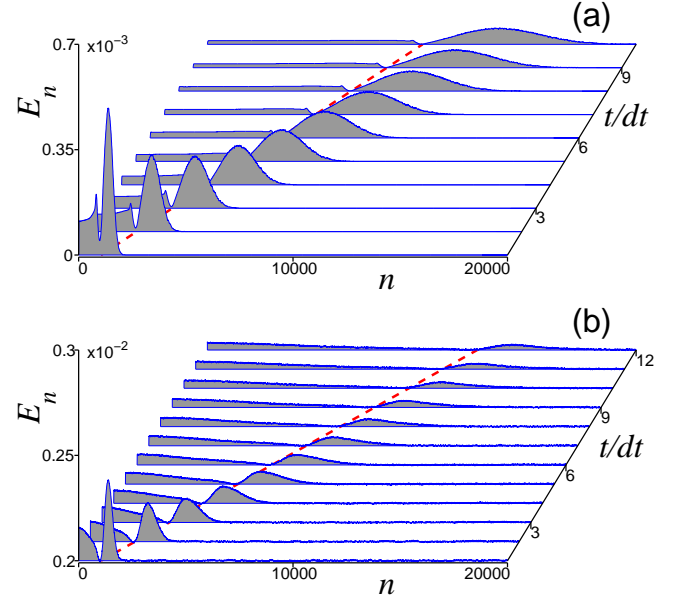


Figure 4: Propagation of energy E_n in the chain with the Lennard-Jones potential (6) for $T_l = 0.01, T_0 = 0.0$ (a) and $T_0 = 0.002$ (b). Energy distributions are shown for characteristic delay time $dt = 1000$, dashed lines show sound cone $n = v_s t / a$.

consider the chain with the same $N = 20000$, when the first $N_l = 40$ atoms in its left end are thermalized at $T_l = 0.01$. As one can see in Fig. 4 (a), for $T_0 = 0$ the energy spreading can be separated into the two parts, which propagate with the sound and supersonic speeds, respectively. The first part corresponds to energy transfer by small-amplitude linear lattice waves (phonons) while the second one corresponds to energy transfer by supersonic kinks (compression acoustic solitons) produced by hard compression component of the LJ interatomic potential [29–31]. For $T_0 = 0$ MSDTED increases quadratically with time, $\langle \Delta x^2 \rangle \sim t^2$, see line 1 in Fig. 5, which corresponds to the ballistic energy propagation in the zero-temperature (nonthermalized) chain.

The picture of energy propagation changes in the thermalized chain. For $T_0 = 0.002$, the part of the initial energy, which propagates with supersonic speed, starts to dissipate in the chain (because the acoustic solitons have finite mean free path in the thermalized LJ chain), and the normal diffusion of thermal energy is established in the chain, see Fig. 4 (b). Time dependence of MSDTED shows that for $t > 5000$ the initial ballistic energy propagation (superdiffusion) is replaced by the normal energy diffusion when $\langle \Delta x^2 \rangle$ grows linearly with time, see line 2 in Fig. 5. In the chain with the combined potential (5) or (7), the anomalous superdiffusion of energy takes place and MSDTED grows as a power function of time: $\langle \Delta x^2 \rangle \propto t^\beta$ with the exponent $1 < \beta < 2$. In the chain with potential (7), $\beta = 1.61$ at $T_0 = 0.002$, see line 3 in Fig. 5; in the chain with potential (5), $\beta = 1.544$ at $T_0 = 0.3$, see line 2 in Fig. 6. Again the minimal delay time, at which the normal energy diffusion is established, is consistent with the minimal

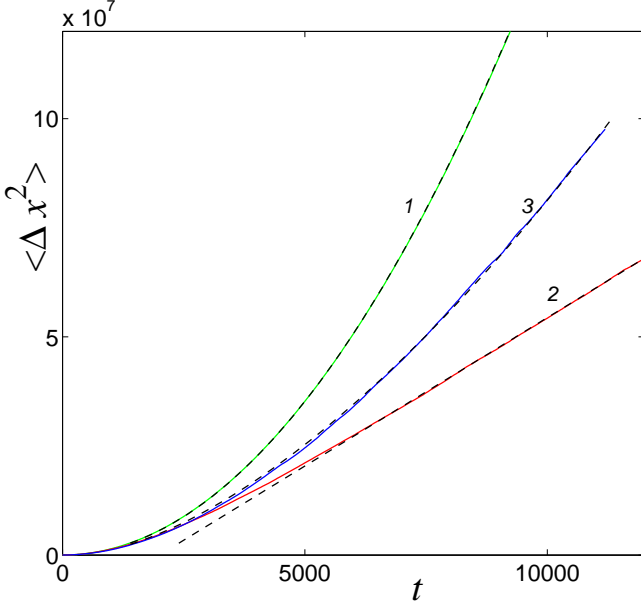


Figure 5: Time dependence of MSDTED, Eq. (10), in the chain with the Lennard-Jones potential (6) with temperature $T_l = 0.01$ of the left end and temperature $T_0 = 0$ and $T_0 = 0.002$ (solid lines 1 and 2) of the rest of the chain, and in the chain with the combined potential (7) for $T_0 = 0.002$, $T_1 = 0.01$ (solid line 3). Dashed lines 1, 2 and 3 give the quadratic $1.41t^2$, linear $6800(t - 2000)$ and power-law $31(t - 300)^{1.61}$ dependencies.

chain length, at which the normal TC is established, cf. Figs. 5 and 6 with Fig. 1.

The value of the exponent β can be related with the exponent α in the length dependence of anomalous TC, when $\kappa \propto N^\alpha$. By the definition of the thermal energy diffusion D_E and TC $\kappa = cD_E$ coefficients, where c is the specific heat density, MSDTED grows as a power function of time as

$$\langle \Delta x^2 \rangle \propto D_E(N)t \propto N^\alpha t, \quad (11)$$

$$\langle \Delta x^2 \rangle \propto t^\beta, \quad (12)$$

where $D_E(N) = \kappa(N)/c \propto N^\alpha$. Below we show that (at least) two different relations between α and β can be obtained from Eqs. (11) and (12) under different assumptions. According to its definition (10), $\langle \Delta x^2 \rangle$ scales as the square of the dimensionless length. Then under the assumption

$$\langle \Delta x^2 \rangle \propto N^2, \quad (13)$$

we get from Eqs. (11) and (12) that

$$\alpha = 2 - 2/\beta. \quad (14)$$

Relation (13) corresponds to the assumption that the regime of the thermal diffusion is reached only in the chain, whose length reaches (or exceeds) the effective phonon mean free path. But under the different assumption

$$t \propto Na/v_s, \quad (15)$$

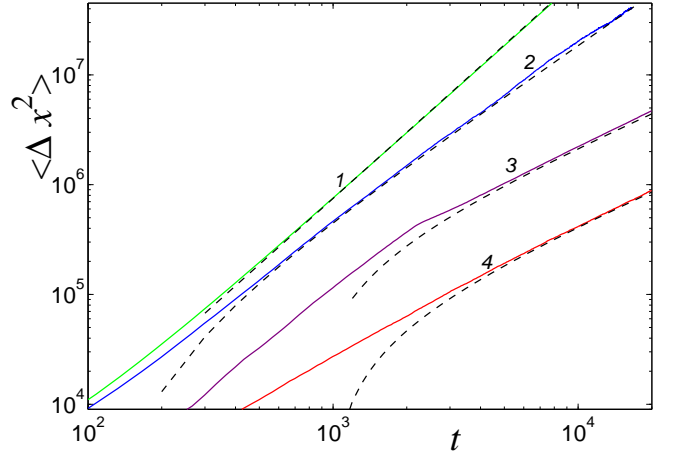


Figure 6: Time dependence of MSDTED, Eq. (10), in the chain with the combined potential (5) with temperature $T_l = 1$ of the left end and temperature $T_0 = 0$ and $T_0 = 0.3$ (solid lines 1 and 2) of the rest of the chain. Dotted lines 1 and 2 give the dependencies $0.7t^2$ and $12.5(t - 110)^{1.544}$. Solid lines 3 and 4 give the dependencies for the chain with the periodic interatomic potential (4) for $T_0 = 0.2$ and $T_0 = 0.3$. Dashed lines 3 and 4 give the dependencies $230(t - 800)$ and $45(t - 964)$.

we get from Eqs. (11) and (12) that

$$\alpha = \beta - 1. \quad (16)$$

Relation (15) corresponds to the assumption that the energy carriers launched from the hotter side of the chain will (ballistically) reach the colder side and reflect back beyond the delay time t . Both scaling relations (14) and (16) imply that normal energy diffusion ($\beta = 1$) leads to the normal (non-divergent) TC ($\alpha = 0$), while the superdiffusion ($\beta > 1$) corresponds to anomalous ($\alpha > 0$) TC. Relation (14) was suggested in Refs. [8, 23], while the relation (16) was obtained in Refs. [24–27] in the specific case of billiard-like 1D models in which non-interacting particles undergo Lévy flights. It is worth noting that the assumption (13) was implicitly used in derivation of Eq. (14) in Ref. [8], see also Ref. [32], and the assumption (15) was explicitly used in derivation of Eq. (16) in Ref. [27]. Moreover, the scaling relation (14) can be derived from the analysis of the diffusion with a position-dependent diffusion coefficient, see, e.g., Ref. [33], under the same assumption (13).

The main conclusion of our analysis is that neither of the relations (14) and (16) is the universal relation, which can be applied to all the nonlinear systems with anomalous heat transport and superdiffusion of thermal energy. We can compare this conclusion with the known conjecture on the absence of the unique velocity-correlation function in turbulent flow, which is universal for all relevant scales and types of flow, see Refs. [34, 35]. Our modeling of anomalous thermal conductivity and superdiffusion of thermal energy in the chains with the combined interatomic potentials (5) and (7) confirms with high accuracy the relation (14): we have $\alpha = 0.705$, $\beta = 1.544$ for the chain with the potential (5), and $\alpha = 0.76$,

$\beta = 1.61$ for the chain with the potential (7). On the other hand, the relation (16) was confirmed in the study of heat transport in the billiard-like 1D system containing colliding particles with two different masses at $T_0 = 0$ [25, 36]. We relate the difference between Eqs. (14) and (16) with the fact that Eq. (14) is applied mostly to the 1D lattices of the coupled (anharmonic) oscillators, in which heat is transported by weakly-scattered *waves* (phonons), while Eq. (16) is applied mostly to the billiard-like 1D systems, in which heat is transported by noninteracting *particles* performing Lévy flights.

In conclusion, we show that the normal thermal conductivity is always accompanied by the normal energy diffusion in the thermalized anharmonic chains, while the superdiffusion of energy is inherent in the thermalized chains with only

anomalous heat transport. We confirm that the confining interparticle potential makes both heat transport and energy diffusion anomalous in low-dimensional phononic systems. We show that the scaling relation between the exponents in time dependence of the mean square displacement of thermal energy distribution and in length dependence of anomalous thermal conductivity is not universal and can be different, depending on the main mechanism of energy transport: either by weakly-scattered waves or by noninteracting colliding particles performing Lévy flights.

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